

Nuclear Spin Diffusion Mediated by Heavy Hole Hyperfine Non-Collinear Interactions

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We show that the hyperfine mediated dynamics of heavy hole states confined in neutral self-assembled quantum dots leads to a nuclear spin diffusion mechanism. It is found that the oftentimes neglected effective heavy hole hyperfine non-collinear interaction is responsible for the low degree of nuclear spin polarization in neutral quantum dots. Moreover, our results demonstrate that after pumping the nuclear spin state is left in a complex mixed state, from which it is not straightforward to deduce the sign of the Ising-like interactions.

We are currently in the midst of an effort to develop reliable nanostructures that can be used to host qubits. Among the possible architectures [1–4], the progress made with spin-based qubits confined in semiconductor structures [5] has been the most impressive [6]. In only a decade, it became possible to efficiently initialize [7], manipulate coherently [8–14], and measure the state of a single spin confined in both electrically defined and self-assembled quantum dots. All of these remarkable achievements are, however, mitigated by poor coherence times on the order of tens of nanoseconds [8–11]. In quantum dots made out of III-V materials, the fluctuations of the nuclear spin felt by the electronic spin through the hyperfine interaction are the main source of decoherence [8, 9, 15–20]. Nevertheless, dynamical decoupling schemes have improved the situation and revealed longer dephasing times [21–24].

From another perspective, nuclear spins are a helpful resource for quantum computing. In gate defined dots, coherent manipulation of electron spin states via the hyperfine interaction has been demonstrated [13, 25, 26]. In self-assembled dots, direct control of nuclear spins has been realized via nuclear magnetic resonance (NMR) [27, 28], which allows to control the direction of the Overhauser field and consequently can be used to control an electron (hole) spin-based qubit. In spite of the efforts made to harness nuclear spins, the role of heavy holes in the dynamics is not yet fully understood.

The first theories suggested an Ising-like type of interaction with a strength on the order of 10% of the one of the electron and with opposite sign [30, 31], which was experimentally verified [32, 33]. However, subsequent experiments seem to contradict these early results. It has recently been claimed that the sign of the coupling strength is opposite for cations and anions [29]. Some other recent experiments [20, 34] report results which indicate a feedback mechanism between heavy holes and nuclear spins. Theories based on p -symmetric Bloch functions for hole states predict that flip-flop terms similar to those of the electronic hyperfine Hamiltonian are very weak [30, 31]. Consequently, it was proposed that non-collinear hyperfine interactions could account for the joint heavy hole nuclear spin dynamics [35, 36]. However,

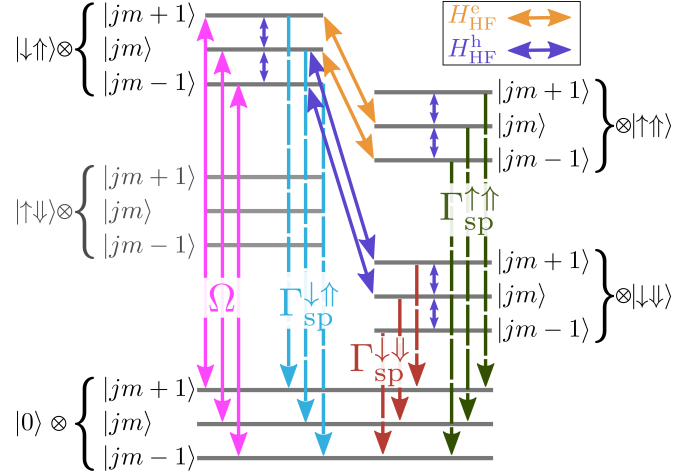


Figure 1. (Color online). Level scheme of the excitonic states in a neutral quantum dot showing optically driven transitions with Rabi frequency Ω under the absorption of σ_+ polarized light (magenta). The nuclear states are described with the total angular momentum j and magnetization m . Hyperfine mediated transitions via the electron are shown in orange and in purple for the hole spin. The excited states relax via spontaneous emission with rates $\Gamma_{sp}^{\uparrow\downarrow} \approx \Gamma_{sp}^{\downarrow\uparrow} \gg \Gamma_{sp}^{\uparrow\uparrow} \approx \Gamma_{sp}^{\downarrow\downarrow}$.

non-collinear interactions were predicted to only affect the dynamics if the laser frequency is not on resonance with the electronic transition which is being driven. An alternative explanation would be that hole states have to be described by both p - and d -type Bloch functions [29] leading to a stronger flip-flop exchange mechanism.

In this letter, by focusing on optical pumping of nuclear spins in neutral quantum dots, we show that the effective hyperfine interaction for heavy hole states, described with p -symmetric Bloch functions, via the non-collinear term leads to an effective nuclear spin diffusion mechanism. Opposite to earlier theories, we find that non-collinear interactions influence the nuclear spin dynamics even when the laser frequency is on resonance with the optically allowed electronic transition. Ironically, nuclear spin diffusion mediated by heavy holes is allowed due to the electron hyperfine interaction which drives the system to a quasi optical dark state. The longer the system stays in the dark state the more efficient diffusion be-

comes. Our results not only provide an explanation for the experimentally observed low degrees of nuclear spin polarization, but they also offer an alternative explanation to the results found in Ref. [29] since the orientation of nuclear spins cannot be assumed to be solely defined by the pumping scheme. Finally, we simultaneously propose a simple experiment aiming at detecting and cancelling the effective heavy hole non-collinear interaction.

The effective Hamiltonian [37],

$$H = H'_0 + H'_L + H_Z^{\text{nuc}} + H_{\text{HF},z}^e + H_{\text{HF},z}^h + H_{\text{HF},\text{nc}}^h, \quad (1)$$

describes the coherent dynamics of the system in the presence of an external magnetic field oriented along the growth axis of the quantum dot (Faraday geometry) and when the laser frequency is close to resonance with the transition $|0\rangle \leftrightarrow |\downarrow\uparrow\rangle$ [c.f. Fig. 1]. The Hamiltonian H'_0 describes the evolution of the exciton states,

$$H'_0 = \frac{\hbar\Delta}{2} (|0\rangle\langle 0| + |\downarrow\uparrow\rangle\langle\downarrow\uparrow|) + \left(\frac{\hbar\Delta}{2} + E_{\downarrow\uparrow}^{\uparrow\downarrow}\right) |\uparrow\downarrow\rangle\langle\uparrow\downarrow| + \left(\frac{\hbar\Delta}{2} + E_{\downarrow\uparrow}^{\uparrow\uparrow}\right) |\uparrow\uparrow\rangle\langle\uparrow\uparrow| + \left(\frac{\hbar\Delta}{2} + E_{\downarrow\uparrow}^{\downarrow\downarrow}\right) |\downarrow\downarrow\rangle\langle\downarrow\downarrow|, \quad (2)$$

where Δ is the laser detuning and we have

$$\begin{aligned} E_{\downarrow\uparrow}^{\uparrow\downarrow} &= -\sqrt{\delta_1^2 + g_+^2 \mu_B^2 B^2}, \\ E_{\downarrow\uparrow}^{\uparrow\uparrow} &= -\delta_0 + \sqrt{\delta_2^2 + g_+^2 \mu_B^2 B^2} - \sqrt{\delta_1^2 + g_-^2 \mu_B^2 B^2}, \\ E_{\downarrow\uparrow}^{\downarrow\downarrow} &= -\delta_0 - \sqrt{\delta_2^2 + g_+^2 \mu_B^2 B^2} - \sqrt{\delta_1^2 + g_-^2 \mu_B^2 B^2}. \end{aligned} \quad (3)$$

Here, we have defined $g_+ = g_e + 3g_h$ and $g_- = g_e - 3g_h$ with g_e (g_h) the electron (heavy hole) Landé g -factor, and μ_B is the Bohr magneton. The coefficients δ_0 , δ_1 , and δ_2 describe respectively the fine structure splitting between bright and dark excitons, among bright, and among dark excitons [38]. Since we are considering σ_+ circularly polarized light and working in a Faraday geometry, the evolution of $|\uparrow\downarrow\rangle$ is trivial. We can therefore reduce the complexity of the problem by omitting this state. The laser Hamiltonian reads

$$H'_L = \hbar\Omega (|0\rangle\langle\downarrow\uparrow| + |\downarrow\uparrow\rangle\langle 0|). \quad (4)$$

where Ω is the Rabi frequency. The nuclear Zeeman Hamiltonian is given by

$$H_n^Z = g_n \mu_n B I_z, \quad (5)$$

with g_n the nuclear Landé g -factor and μ_n the nuclear Bohr magneton. The electronic hyperfine Hamiltonian within the homogeneous coupling approximation is given by,

$$H_{\text{HF}}^e = H_{\text{HF},z}^e + H_{\text{HF},\perp}^e = A^e \left(S_z I_z + \frac{1}{2} (S_+ I_- + S_- I_+) \right). \quad (6)$$

Here, S_z (I_z) is the electron (nuclear) spin operator in z direction and we have introduced the ladder operators, $S_{\pm} = S_x \pm iS_y$ and $I_{\pm} = I_x \pm iI_y$. We denote the average hyperfine coupling constant as A^e , the longitudinal part of Eq. (6) by $H_{\text{HF},z}^e$, and the transverse one by $H_{\text{HF},\perp}^e$. The effective hyperfine Hamiltonian for heavy holes can be written as [37],

$$\begin{aligned} H_{\text{HF}}^h &= H_{\text{HF},z}^h + H_{\text{HF},\perp 1}^h + H_{\text{HF},\perp 2}^h + H_{\text{HF},\text{nc}}^h \\ &= A_z^h S_z I_z + A_{\perp 1}^h S_+^h I_- + A_{\perp 1}^{h*} S_-^h I_+ \\ &\quad + A_{\perp 2}^h S_+^h I_+ + A_{\perp 2}^{h*} S_-^h I_- + A_{\text{nc}}^h S_z^h I_+ + A_{\text{nc}}^{h*} S_z^h I_-, \end{aligned} \quad (7)$$

where S_i^h ($i = z, \pm$) are pseudospin operators for the effective heavy hole states [37]. We use a similar notation to the one introduced in Eq. (6) for longitudinal and transverse interactions. We denote the non-collinear term by $H_{\text{HF},\text{nc}}^h$.

Here, we follow the procedure of Refs. [39, 40] and describe the contribution of the transverse (flip-flop) terms of Eqs. (6) and (7) on the dynamics as a dissipative process. The evolution of the system is then described by the Lindblad master equation [41], $\dot{\rho} = -\frac{i}{\hbar}[H, \rho] + \sum_{j=1}^{d^2-1} ([L_j \rho, L_j^\dagger] + [L_j, \rho L_j^\dagger])/2$, with d the dimension of the Hilbert space. Since we only consider dissipative processes within the electronic subspace, we get by with less Lindblad operators. We take into account spontaneous emission from the bright exciton $|\downarrow\uparrow\rangle$ and from both (quasi) dark excitons to the ground state [29].

These are respectively described by $L_1 = \sqrt{\Gamma_{\text{sp}}^{\downarrow\uparrow}} |0\rangle\langle\downarrow\uparrow|$, $L_2 = \sqrt{\Gamma_{\text{sp}}^{\uparrow\downarrow}} |0\rangle\langle\uparrow\downarrow|$, and $L_3 = \sqrt{\Gamma_{\text{sp}}^{\downarrow\downarrow}} |0\rangle\langle\downarrow\downarrow|$, where Γ_{sp}^j , $j = \downarrow\uparrow, \uparrow\downarrow, \downarrow\downarrow$, is the spontaneous decay rate. We describe the nuclear spin state with its total angular momentum j and its projection along the magnetic field given by m [c.f. Fig. 1]. The Lindblad operators $L_4 = \sqrt{\Gamma_e^{\downarrow\uparrow}} |0, j, m-1\rangle\langle\downarrow\uparrow, j, m|$ and $L_5 = \sqrt{\Gamma_h^{\downarrow\uparrow}} |0, j, m+1\rangle\langle\downarrow\uparrow, j, m|$ respectively describe electron and hole flip-flop processes. The rates $\Gamma_e^{\downarrow\uparrow}$ and $\Gamma_h^{\downarrow\uparrow}$ are calculated with the same method as in Ref. [40], we find

$$\Gamma_e^{\downarrow\uparrow} \simeq \frac{\Gamma_{\text{sp}}^{\uparrow\downarrow}}{4} \left| \frac{A^e \sqrt{j(j+1) - m(m-1)}}{E_{\uparrow\downarrow}^{\downarrow\uparrow} - A^e(m - \frac{1}{2}) + \frac{3}{2}A_z^h + g_n \mu_n B} \right|^2, \quad (8)$$

where we have neglected the contribution coming from $H_{\text{HF},\perp 2}^h$ since $|A_{\perp 2}^h|/A^e \ll 1$ and

$$\Gamma_h^{\downarrow\uparrow} = \frac{\Gamma_{\text{sp}}^{\downarrow\downarrow}}{4} \left| \frac{A_{\perp 1}^h \sqrt{j(j+1) - m(m+1)}}{E_{\downarrow\downarrow}^{\downarrow\uparrow} + \frac{1}{2}A^e + 3A_z^h(m + \frac{1}{2}) - g_n \mu_n B} \right|^2. \quad (9)$$

We assume nuclear spins to be initially in a thermal state. This is a reasonable assumptions even for experiments performed at low temperatures, where the thermal energy is larger than the nuclear Zeeman, $k_B T \gg E_Z^{\text{nuc}}$, with k_B the Boltzmann's constant. Thus, at $t = 0$, the

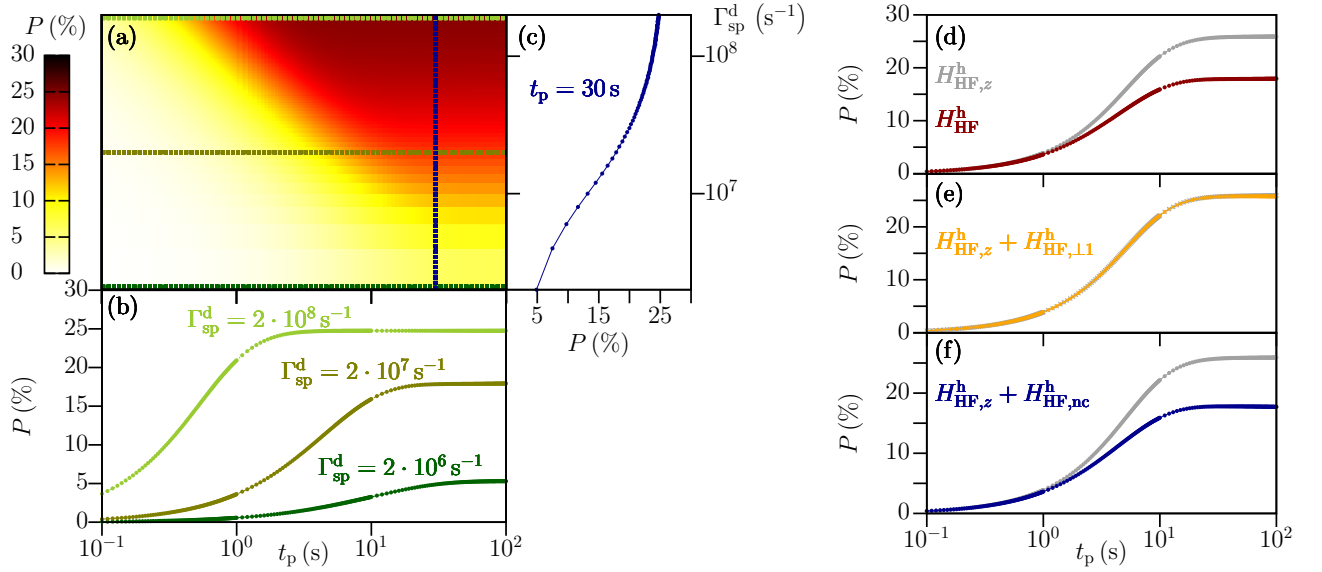


Figure 2. (Color online). (a) Nuclear spin polarization P as a function of pumping time t_p and spontaneous decay rate $\Gamma_{\text{sp}}^{\uparrow\uparrow} \approx \Gamma_{\text{sp}}^{\downarrow\downarrow} = \Gamma_{\text{sp}}^{\text{d}}$. Values of other parameters are given in the main text. (b) Traces taken along $\Gamma_{\text{sp}}^{\text{d}} = 2 \cdot 10^8, 2 \cdot 10^7$, and $2 \cdot 10^6$ Hz. P saturates at lower values for smaller $\Gamma_{\text{sp}}^{\text{d}}$'s. (c) Trace taken along $t_p = 30$ s showing the dependence on $\Gamma_{\text{sp}}^{\text{d}}$. (d) Comparison of P as a function of t_p between $H_{\text{HF}}^{\text{h}} = H_{\text{HF},z}^{\text{h}}$ (gray), for which saturation corresponds to formation of a nuclear spin dark state, and the full effective Hamiltonian Eq. (7) (red) for $\Gamma_{\text{sp}}^{\text{d}} = 2 \cdot 10^7$ Hz. (e) Same as (d) but compared with $H_{\text{HF}}^{\text{h}} = H_{\text{HF},z}^{\text{h}} + H_{\text{HF},\perp 1}^{\text{h}}$. The result shows that heavy hole mediated flip-flop processes are negligible. (f) Same as (d) but compared with $H_{\text{HF}}^{\text{h}} = H_{\text{HF},z}^{\text{h}} + H_{\text{HF},\text{nc}}^{\text{h}}$. The heavy hole hyperfine non-collinear interaction is the origin of the lower values of P since it leads to an effective nuclear spin diffusion mechanism.

nuclear spins are assumed to be in a fully mixed state. Further assuming spin-1/2 for the nuclei, we have [40]

$$\rho_{\text{nuc}} = \sum_{j,m} \frac{(2j+1)N! [\Theta(j+m) - \Theta(j-m)]}{(\frac{N}{2} + j + 1)! (\frac{N}{2} - j)! 2^N} |j, m\rangle \langle j, m|, \quad (10)$$

with N the number of nuclear spins and $\Theta(x)$ is the Heaviside theta function. The initial electronic state is given by the quantum dot vacuum, i.e. $\rho_e = |0\rangle\langle 0|$. Thus, the density matrix describing the whole system at $t = 0$ is written as $\rho = \rho_e \otimes \rho_{\text{nuc}}$.

In Fig. 2(a), we present the degree of nuclear spin polarization $P = \text{Tr}[\rho_{\text{nuc}}(t_p)I_z]/P_{\text{max}}$ as a function of pumping time t_p and spontaneous decay rate of the dark states $\Gamma_{\text{sp}}^{\text{d}}$, where $P_{\text{max}} = N/2$. Since the ratio of the dark states energy is nearly one, $E_{\uparrow\uparrow}/E_{\downarrow\downarrow} \approx 1$ [37], we have $\Gamma_{\text{sp}}^{\uparrow\uparrow} \simeq \Gamma_{\text{sp}}^{\downarrow\downarrow} = \Gamma_{\text{sp}}^{\text{d}}$. The calculations were performed with $\delta_0 = 5.6213 \cdot 10^{11}$ Hz, $\delta_1 = \delta_2 = 5.3174 \cdot 10^{10}$ Hz, $B = 8$ T, $g_e = -0.35$, $g_h = 0.63$, $g_n \mu_n = 3.3 \cdot 10^{-8}$ eV/T, $\Delta = 0$ Hz, $\Omega = 2.03 \cdot 10^{10}$ Hz, $A^e = 10^8$ Hz, $A_z^h = -10^7$ Hz, $|A_{\perp 1}^h| = 3 \cdot 10^5$ Hz, $A_{\text{nc}}^h = 3 \cdot 10^5$ Hz, $\Gamma_{\text{sp}}^{\downarrow\uparrow} = 2 \cdot 10^9$ Hz, and $N = 30$. We have cancelled out the imaginary part of $H_{\text{HF},\text{nc}}^{\text{h}}$ by performing a suitable rotation of angle θ , $U = \exp[i\theta I_z]$. We notice that the saturation of the nuclear polarization depends strongly on the lifetime of the dark states. To demonstrate clearly this behavior, we present traces taken respectively for different values of $\Gamma_{\text{sp}}^{\text{d}}$ [Fig. 2(b)] and for $t_p = 30$ s [Fig. 2(c)], for which P

has reached saturation. Both of these traces show that P saturates at smaller values for slower $\Gamma_{\text{sp}}^{\text{d}}$. To come to a clear mechanism that explains this result, we compare P as function of t_p for $\Gamma_{\text{d}} = 10^7 \text{ s}^{-1}$ between an Ising-like ($H_{\text{HF},z}^{\text{h}}$) and other forms of the effective heavy hole hyperfine Hamiltonian. In Fig. 2(d), we compare P obtained with $H_{\text{HF},z}^{\text{h}}$ (gray) to the effective Hamiltonian given by Eq. (7) (red). The different values of P at saturation indicate that the relatively small corrections to the Ising-like hyperfine Hamiltonian influence the dynamics. For $H_{\text{HF},z}^{\text{h}}$, P saturates due to formation of a nuclear spin dark state [40]. To tell apart the contribution of $H_{\text{HF},\perp 1}^{\text{h}}$ and $H_{\text{HF},\text{nc}}^{\text{h}}$, we plot P obtained with $H_{\text{HF},z}^{\text{h}}$ (gray) and $H_{\text{HF},z}^{\text{h}} + H_{\text{HF},\perp 1}^{\text{h}}$ (orange) in Fig. 2(e), which show that heavy hole flip-flop processes are irrelevant for the nuclear spin dynamics. In the presence of a large magnetic field and due to the smallness of $|A_{\perp 1}^h|$, the heavy hole forbidden relaxation rate $\Gamma_{\text{h}}^{\downarrow\uparrow}$ is too slow compared to electron forbidden relaxation rate $\Gamma_{\text{e}}^{\downarrow\uparrow}$ and to spontaneous emission $\Gamma_{\text{sp}}^{\downarrow\uparrow}$ to have an impact on the dynamics. Finally, we verify that the non-collinear interaction is responsible for saturations lower than the nuclear dark state limit. In Fig. 2(f), we show P calculated with $H_{\text{HF},z}^{\text{h}}$ (gray) and $H_{\text{HF},z}^{\text{h}} + H_{\text{HF},\text{nc}}^{\text{h}}$ (blue). Our results demonstrate that the heavy hole non-collinear hyperfine interaction leads to an effective nuclear spin diffusion mechanism that hin-

ders P . As it can be observed from Figs. 2(a), (b), and (c), the diffusion becomes more prominent when the system is held for a relatively long time in one of the optical dark states. It has been reported that the oscillator strength for optical dark states is a hundred to a thousand times smaller than the oscillator strength of bright states [29], which implies $\Gamma_{\text{sp}}^{\downarrow\uparrow}/\Gamma_{\text{sp}}^{\text{d}} \approx 100 - 1000$. Finally, our results indicate that upon reaching saturation most of the nuclear spin states are still populated and the system is left in a mixed state. Thus, our findings suggest that there could be an alternative interpretation of recent experimental results about the sign of the Ising-like interaction [29]. The unexpected shift of the Overhauser field could simply originate from nuclear spin diffusion, which lowers P , when measuring the spectral position of the optical dark states.

In the following, we propose a simple experiment to detect and simultaneously cancel the presence of non-collinear interactions. The idea is to change the orientation of the external magnetic field to transform the nuclear Zeeman Hamiltonian into, $H_n^Z = g_n \mu_n B \cos(\varphi) I_z + g_n \mu_n B \sin(\varphi)(I_+ + I_-)/2$, with φ the rotation angle. In our coordinate system the magnetic field has to be rotated around the y -axis, i.e. φ is the angle between the z -axis and \mathbf{B} . We solve again a Lindblad master equation, but with a Hamiltonian that takes into account that \mathbf{B} is not necessarily aligned with the growth axis of the quantum dot. In addition to the trivial change $B \rightarrow B \cos(\varphi) \equiv B_z$ in Eqs. (1), (8), and (9) as well as the discussed modification of the nuclear Zeeman Hamiltonian, we also need to take into account that misalignment of \mathbf{B} leads to mixing of bright and dark excitons via $H_{\text{bd}} = g_e \mu_B B \sin(\varphi)(S_+ + S_-)/4 + g_h^{xx} \mu_B B \sin(\varphi)(S_+^h + S_-^h)/4$, with $g_h^{xx} \simeq g_h/10$ [42, 43] the heavy hole Landé g -factor along the x -axis. We also add to the dissipative part of the Lindblad equation spontaneous relaxation from $|\uparrow\downarrow\rangle$ to the ground state with rate $\Gamma_{\text{sp}}^{\uparrow\downarrow}$ and two non-conserving nuclear spin relaxation mechanisms. These are described by $L_6 = \sqrt{\Gamma_{\text{sp}}^{\uparrow\downarrow}}|0\rangle\langle\uparrow\downarrow|$, $L_7 = \sqrt{\Gamma_{\text{e}}^{\uparrow\downarrow}}|0, j, m+1\rangle\langle\uparrow\downarrow, j, m|$, and $L_8 = \sqrt{\Gamma_{\text{h}}^{\uparrow\downarrow}}|0, j, m-1\rangle\langle\uparrow\downarrow, j, m|$ with

$$\Gamma_{\text{e}}^{\uparrow\downarrow} \simeq \frac{\Gamma_{\text{sp}}^{\uparrow\downarrow}}{4} \left| \frac{A^{\text{e}} \sqrt{j(j+1) - m(m+1)}}{E_{\uparrow\downarrow}^{\uparrow\downarrow} + A^{\text{e}}(m + \frac{1}{2}) + \frac{3}{2}A_z^{\text{h}} - g_n \mu_n B_z} \right|^2, \quad (11)$$

and

$$\Gamma_{\text{h}}^{\uparrow\downarrow} = \frac{\Gamma_{\text{sp}}^{\uparrow\downarrow}}{4} \left| \frac{A_{\uparrow\downarrow}^{\text{h}} \sqrt{j(j+1) - m(m-1)}}{E_{\uparrow\downarrow}^{\uparrow\downarrow} + \frac{1}{2}A^{\text{e}} + \frac{3}{2}A_z^{\text{h}}(m - \frac{1}{2}) + g_n \mu_n B_z} \right|^2. \quad (12)$$

In Fig. 3(a), we plot the nuclear spin polarization P as a function of t_p and φ . We use the same set of parameters as before and $\Gamma_{\text{sp}}^{\text{d}} = 10^7 \text{ s}^{-1}$. As for the optical dark state, we have $E_{\downarrow\uparrow}/E_{\uparrow\downarrow} \approx 1$ which allows us to write $\Gamma_{\text{sp}}^{\uparrow\downarrow} \simeq \Gamma_{\text{sp}}^{\downarrow\uparrow} = \Gamma_{\text{sp}}^{\text{b}}$. The results show that

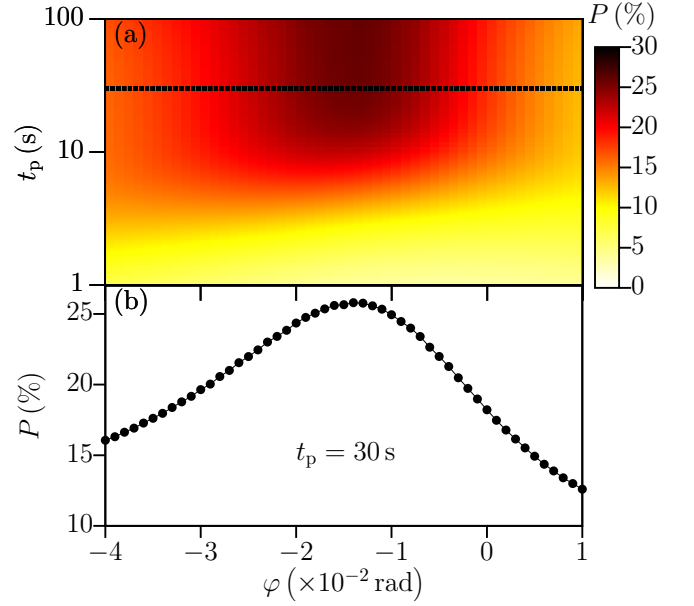


Figure 3. (Color online). (a) Nuclear spin polarization P as a function of t_p and φ (angle between the external magnetic field \mathbf{B} and the z -axis) for $\Gamma_{\text{sp}}^{\text{d}} = 2 \cdot 10^7 \text{ s}^{-1}$. (b) Trace taken along $t_p = 30$ s. The effect of the non-collinear interaction can be cancelled by orienting the magnetic field opposite to the effective Overhauser field defined by the hyperfine non-collinear Hamiltonian.

the non-collinear heavy hole hyperfine interaction is fully cancelled at $\varphi \simeq -0.014$ rad, for which we retrieve the saturation limit set by the nuclear spin dark state. In Fig. 3(b), we show a trace taken for $t_p = 30$ s.

In conclusion, we have shown that the effective heavy hole hyperfine interaction via non-collinear terms influences nuclear spin dynamics. In particular, we have shown how to experimentally detect and cancel the effects of such interaction. We expect the described effects to be stronger when considering an inhomogeneous hyperfine Hamiltonian since the statistical weight of the states contributing the most to P are not suppressed [40]. Moreover, when trying to cancel the heavy hole non-collinear interaction, a series of maximums should be observed as a function of the rotation angles. Each maximum corresponds to a different nuclear species. This also implies that none of the maximums correspond to the limit set by the formation of a nuclear dark state.

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